Response to Comments of Reviewer #2

Manuscript number: acp-2021-611
Title: Simulated impacts of vertical distributions of black carbon aerosol on meteorology and $PM_{2.5}$ concentrations in Beijing during severe haze events

General comments:
In the current work, the authors used WRF-Chem with IPR analysis and sensitivity tests to assess the effect of vertical distribution of BC on meteorology and surface $PM_{2.5}$ concentrations during Beijing haze episodes. The research topic is very well suited to the scope of the current journal and the results presented are very interesting to their readers. However, there are some uncertainties, especially in the methodology, that require some modification before it is accepted. Without a concrete explanation of the methodology, the reviewer is uncertain whether the model settings chosen by the author will actually answer the authors' research questions. See the general and specific comments listed below.
Thanks to the referee for the helpful comments and suggestions. We have revised the manuscript carefully and the point to point responses are listed below.

Major concerns/questions:
1. There are no presentations on clouds and precipitation throughout the manuscript. In the presence of clouds and precipitation, the radiation effect of aerosols is quite different from that of sunny days. Please show and compare measured and simulated clouds, precipitations, or solar radiation.

Response:
The comparison between simulated and observed hourly precipitation (mm), 3-hourly total cloud cover (%) and 3-hourly shortwave downward radiation flux (SWDOWN, W m^{-2}) in Beijing are added in Figures 5f-5h (see below). The corresponding statistical metrics are added in Table 3 (see below). We have also added the following sentences in the first paragraph of Section 3.2 to describe the comparison.

'The simulated SWDOWN in CTRL experiment agrees well with the observations with R of 0.76 and MB of -14.9 W m^{-2}. Due to the limitation of the model outputs, the model provides only information of whether there is cloud in the grid or not. The model can reproduce well the presence of cloud during 11-19 December 2016. Both observations and model results show no precipitation in the studied time period.'.

Table 3. Statistical metrics for temperature at 2 m (T2; °C), relative humidity at 2 m (RH2; %), wind speed at 10 m (WS10; m s^{-1}), wind direction at 10 m (WD10, °), PBLH (m), SWDOWN (W m^{-2}), $PM_{2.5}$ (μg m^{-3}), SO_2 (ppbv), NO_2 (ppbv), CO (ppmv) and O_3 (ppbv).

Variables	SIM[a]	OBS[b]	R[c]	MB[d]	NMB[e]	MFB[f]
T2 (°C)	-0.5	-0.6	0.77	0.1	-17.8%	-13.1%
RH2 (%)	52.5	55.8	0.75	-3.4	-6.0%	-0.3%

WS10 (m s^{-1})	1.8	2.3	0.52	-0.5	-20.6%	-11.5%
WD10 (°)	165.6	182.0	0.45	-16.4	-9.0%	0.7%
PBLH (m)	205.8	174.9	0.72	30.9	17.7%	72.9%
SWDOWN (W m^{-2})	86.0	100.8	0.76	-14.9	-14.8%	-17.4%
$PM_{2.5}$ (μg m^{-3})	145.6	132.3	0.77	13.2	10.0%	15.7%
SO_2 (ppbv)	7.9	7.8	0.38	0.1	1.4%	-2.9%
NO_2 (ppbv)	47.7	39.2	0.78	8.5	21.6%	20.2%
CO (ppmv)	1.8	1.9	0.73	-0.1	-4.9%	6.4%
O_3 (ppbv)	6.7	6.8	0.66	-0.1	-1.2%	-36.0%

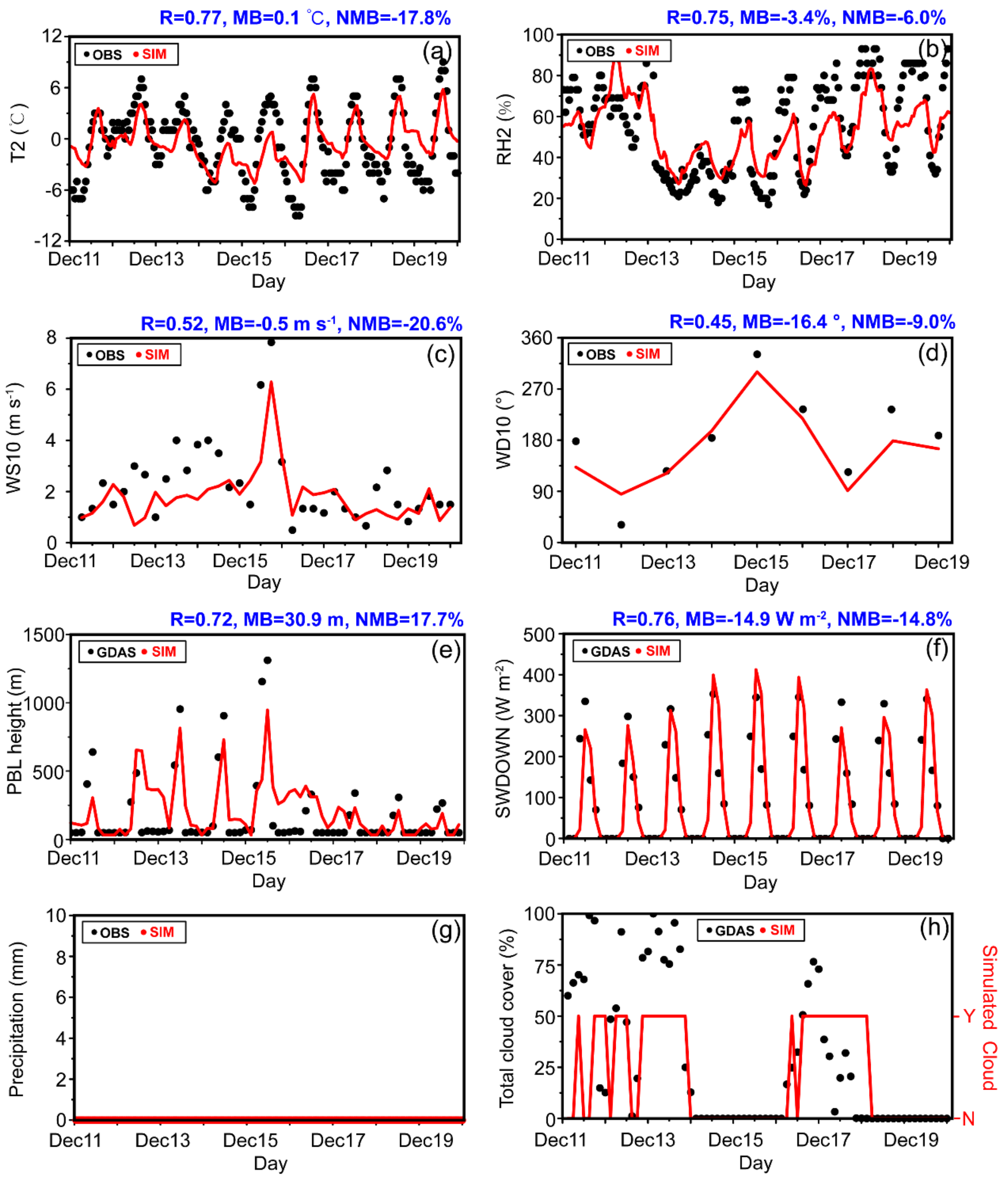

Figure 5. Comparisons of simulated meteorological parameters from CTRL simulation with measurements. The black dots and red lines are the observed (reanalysis) and

simulated hourly data of T2 (°C), RH2 (%), precipitation (mm), and 3-hourly data of PBL height (m), SWDOWN (W m^{-2}), total cloud cover (%), 6-hourly data of WS10 (m s^{-1}), and daily data of WD10 (°) in Beijing from 11 December 2016 to 19 December 2016. PBLH, SWDOWN, and total cloud cover are taken from GDAS. The WRF-Chem model output shows only a grid has cloud (Y) or no cloud (N).

Specific comments:

1. Abstract:
   The authors simply repeated "$PM_{2.5}$ concentration", but more specifically, "$PM_{2.5}$ surface air concentrations" ($PM_{2.5}$ can be aloft). Please define "$PM_{2.5}$ concentration" as "surface air concentrations of $PM_{2.5}$" when it is first appeared.

Response:

We have clarified in the abstract that we mean the surface-layer $PM_{2.5}$ (covers from 0 m to 79.5 m).

2. P. 8, Ln. 154, "NCEP": Please be more specific. For example, specify the datasets number.

Response:

We have added the following details of NCEP in the first paragraph of Section 2.1:

'Meteorological initial and boundary conditions in this model were derived from National Center for Environmental Prediction (NCEP) Final (FNL) Operational Model Global Tropospheric Analyses (ds083.2) with a spatial resolution of 1°×1°.'.

3. P. 9, Ln. 168, "FINN": Please provide horizontal and temporal resolution.

Response:

We have added the following details of FINN in the second paragraph of Section 2.1:

'Biomass burning emissions were taken from the Fire INventory from NCAR (FINNv1.5) which provides daily emissions at a horizontal resolution of ~1 km^2 (Wiedinmyer et al., 2011).'.

4. P. 10, Ln. 189, "updrafts": The convection scheme also includes downdrafts and precipitation. Which parameterization did you use for convection? Please list it in Table 1. The subgrid-scale wet deposition is calculated in the convection model so they can be counted as CONV here, but they need to be count as WET (wet deposition).

Response:

We have revised this sentence as 'CONV refers to the transport within the sub-grid wet convective updrafts, downdrafts and precipitation (Chen et al., 2019).'. We used Grell 3-D scheme (an improved version of the GD scheme) for convection. We have also added it in Table. 1 (see below).

Table1. Physical and chemical options for WRF/Chem.

WRF/Chem Model Configuration	Description

Microphysics scheme	Lin microphysics scheme (Wiedinmyer et al., 2011)
Longwave radiation scheme	RRTMG scheme (Zhao et al., 2011)
Shortwave radiation scheme	RRTMG scheme (Zhao et al., 2011)
Gas phase chemistry scheme	CBMZ (Zaveri and Peters, 1999)
Aerosol module	MOSAIC (Zaveri et al., 2008)
Photolysis scheme	Fast-J (Wild et al., 2000)
Boundary layer scheme	Yonsei University Scheme (YSU) (Hong et al., 2006)
Pavement parameterization scheme	Noah Land Surface Model scheme
Cumulus option	Grell 3-D ensemble scheme

5. P. 10, Ln. 192, "cloud": How do you separate cloud chemical formation from in-cloud scavenging? The formation of $PM_{2.5}$ due to cloud chemistry occurs only when the cloud and rain droplets are completely evaporated. On the other hand, $PM_{2.5}$ in the cloud and rain droplets are not counted as $PM_{2.5}$ and are removed from the air (although they are not completely removed unless droplets reach the ground).

Response:

In the WRC-Chem model, the cloud chemistry is an independent module. We quantify the changes in $PM_{2.5}$ before and after calling this module as the impact of cloud chemistry on $PM_{2.5}$. In-cloud scavenging is included in wet scavenging (WET). WET considers the wet removal of aerosols by in-cloud scavenging and below-cloud washout.

6. P. 10, Ln. 194, "WET represents the wet removal processes of aerosols": In-cloud and below-cloud? Again, how do you separate "cloud" in CHEM from in-cloud scavenging in WET here?

Response:

See our response to your previous question.

7. P. 10, Ln. 195, "OTHER": What are they? Dry deposition should be the one but any other processes?

Response:

OTHER is calculated by subtracting the six physical/chemical processes of $PM_{2.5}$ (CONV, VMIX, CHEM, TRA, WET and EMI) from the total $PM_{2.5}$.

8. P. 10, Ln. 202: "Beijing station", where is it and what does it belong to? Does the station belong to NOAA? Maybe not, but the data was obtained from NOAA's website.

Response:

'Beijing station' is 'Beijing-Capital International Airport station (station number: 54511099999)' in NOAA. We have changed 'Beijing station' to 'Beijing-Capital International Airport station (40.08°N, 116.58°E)' in the revised manuscript. The station (54511099999) belongs to NOAA and it was obtained from the Integrated Surface Dataset (Global).

9. P. 10, Ln. 205, PBL of GDAS: what is the horizontal resolution of GDAS? Even

though the GDAS is the analysis (or one can call it observation), is their PBL also assimilated with observed PBL? If not, PBL of GDAS cannot be regarded as observation as you show in Fig. 5. If it is assimilated with observed PBL, please specify which observation data was assimilated.

Response:

The Global Data Assimilation System (GDAS) has a horizontal resolution of 1°×1°. GDAS continuously collects observational data from the Global Telecommunications System (GTS) and other sources. The PBL is calculated by the meteorological data in GDAS. We have added extra notes in the first paragraph of Section 2.3 : 'Due to the limited observations of planetary boundary layer heights (PBLH), shortwave downward radiation flux (SWDOWN) and total cloud cover in Beijing, the reanalysis data of 3-hourly PBLH, SWDOWN and total cloud cover for Beijing from the Global Data Assimilation System (GDAS) with a spatial resolution of 1°×1° (http://ready.arl.noaa.gov/READYamet.php) were used for model evaluation. More details about the GDAS dataset can be found in Rolph (2013) and Kong et al. (2015).'. We have also changed 'OBS' to 'GDAS' in three panel of PBLH, SWDOWN and total cloud cover in Fig. 5 (see below).

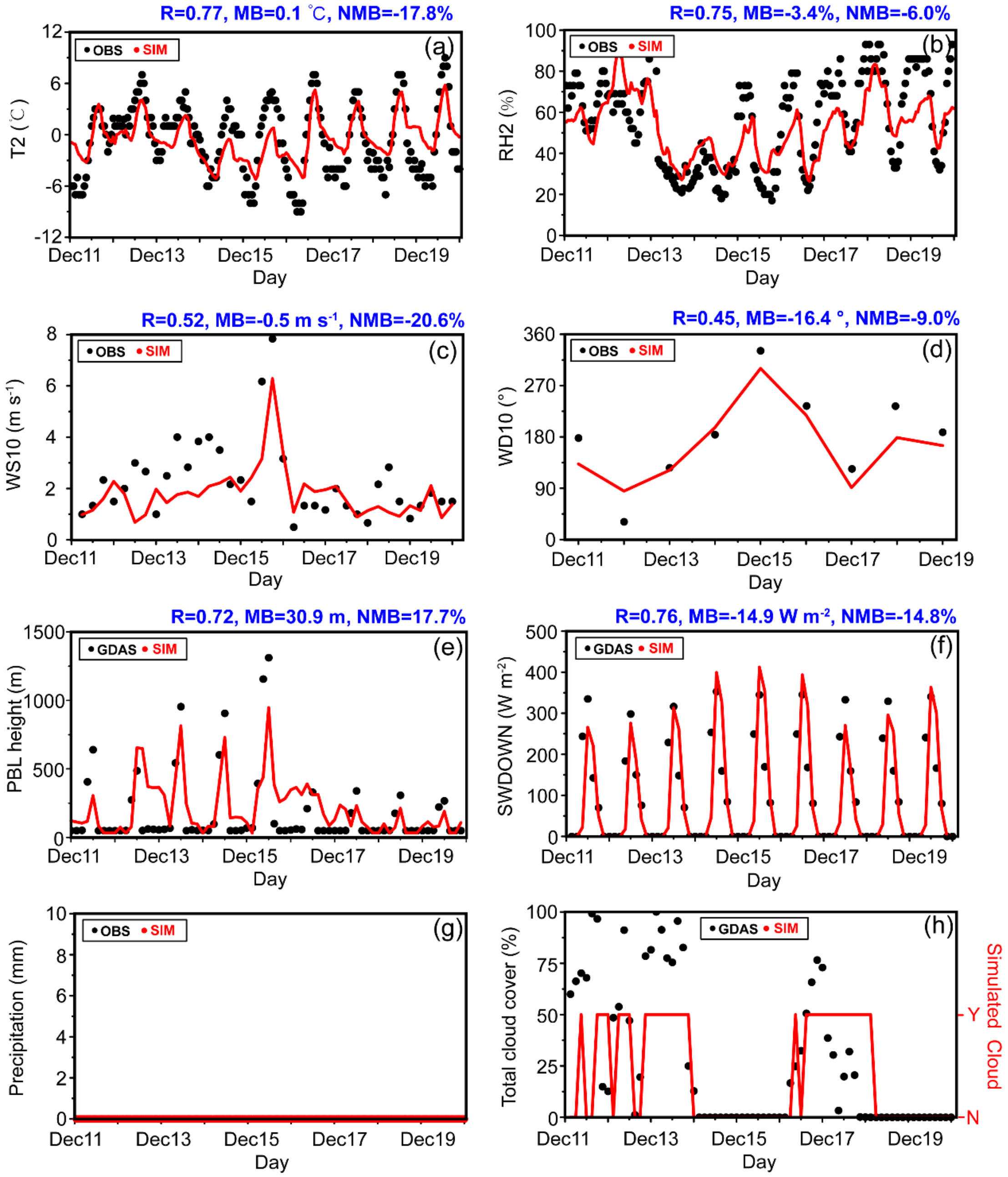

Figure 5. Comparisons of simulated meteorological parameters from CTRL simulation with measurements. The black dots and red lines are the observed (reanalysis) and simulated hourly data of T2 (°C), RH2 (%), precipitation (mm), and 3-hourly data of PBL height (m), SWDOWN (W m^{-2}), total cloud cover (%), 6-hourly data of WS10 (m s^{-1}), and daily data of WD10 (°) in Beijing from 11 December 2016 to 19 December 2016. PBLH, SWDOWN, and total cloud cover are taken from GDAS. The WRF-Chem model output shows only a grid has cloud (Y) or no cloud (N).

10. P. 12, "indirect radiative effects": how? The authors used the Lin's scheme for cloud microphysics, which is a single moment scheme and thus cloud albedo and cloud lifetime effects are not considered. Is it intended simulation settings?

Response:

To run with indirect radiative effects, we turn on the aerosol direct effect and select Lin's microphysics scheme. Then we turn on the prognostic number density option to allow the Lin's scheme to be double moment and be able to communicate the desire to run indirect effect. Such method is based on WRF-Chem User's Guide (https://ruc.noaa.gov/wrf/wrf-chem/user-support.htm).

11. P. 15-16, discrepancy of vertical profiles on December 11: Are the observation and simulation average times same? The simulated profile appears to be at night or very stable during the day, but the observed profile looks only be during the day.

Response:

We have clarified here that 'The observed BC vertical profile is at 16:20 LT and the simulated BC vertical profile is an average of 16:00 and 17:00 LT on December 11.'

12. It is necessary to discuss the reason for the difference in vertical profile between the simulation and observation on December 11th. Judging from the profile, the simulated surface air concentration is four times the observed value, but the overestimation of the simulated surface $PM_{2.5}$ concentration is not so high (Fig. 3). The simulated night T2 of the day has a significant overestimation (+6 deg C). It is 0 deg C in the simulation and -6 deg C in the observation. Is it due to overprediction of simulated clouds to prevent radiative cooling at night?

Response:

We have added the following sentences in the third paragraph of Section 3.1 to discuss the reason for the difference in BC vertical profile between observation and simulation on December 11:

'Possible reasons for model's failing to represent BC vertical profile on December 11 are as follows: (1) the model cannot capture the wind at high altitudes and does not reproduce the high-altitude BC concentrations in the surrounding areas of Beijing; (2) the model underestimates the daily maximum PBLH in Beijing which inhibits the upward transport of surface-layer BC.'

Fig. 5h shows that there was no cloud in the model at night (0:00-8:00 LT) on December 11, so the overestimation of T2 might be caused by the overestimation of surface-layer BC concentrations in the model. The surface-layer BC can absorb solar radiation during daytime and warm the air temperature at night (Ding et al., 2019).

13. Caption of Fig. 1, "The BC vertical profiles were modified for the red box which covers …" should be written in the main text.

Response:

We have added the following sentence here: 'The BC vertical profiles were only modified in the blue square shown in Fig. 1a.' in the last paragraph of Section 2.4.

14. Fig. 2, "blue and red squares" are hardly legible.

Response:

We have changed blue and red squares to black and blue squares (see below).

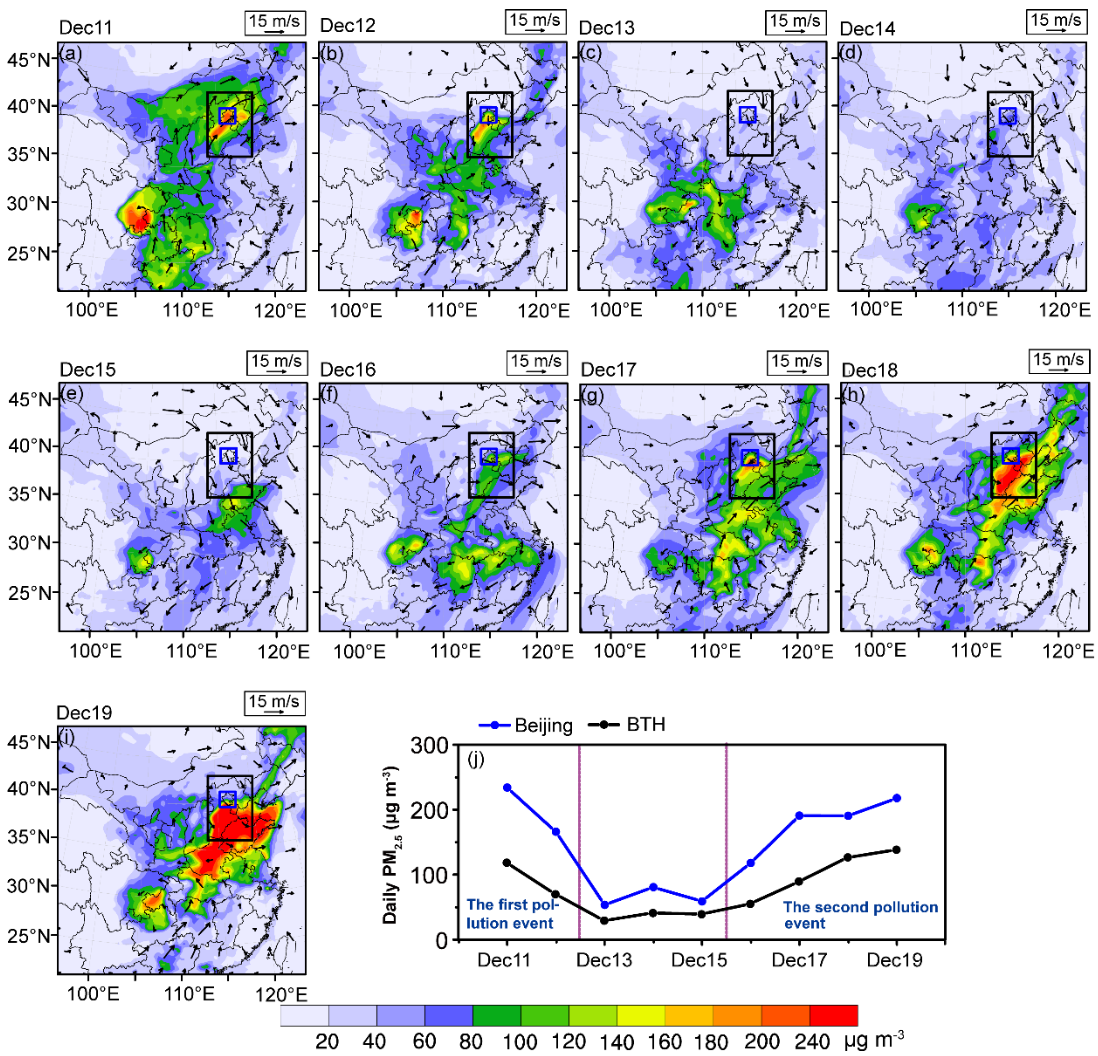

Figure 2. (a-i) Simulated spatial distributions of surface-layer $PM_{2.5}$ concentrations (μg m^{-3}) and winds (m s^{-1}) at 850 hPa at 2 pm local time from 11 to 19 December 2016. Black and blue squares in each panel denote the regions of Beijing-Tianjin-Hebei and Beijing, respectively. (j) Time series of simulated mean daily $PM_{2.5}$ concentration from 11 to 19 December 2016 averaged over the Beijing (blue square) and BTH (black square).

15. Fig. 2(j), Does "Beijing" mean spatial average of the blue square region? Or one grid of the center of Beijing region? Please specify. Throughout the manuscript, it is hard to get whether the authors indicate values of only one grid point, one observation site, or those of spatial average.

Response:

We have clarified this in the figure caption (Fig. 2). 'Time series of simulated daily mean $PM_{2.5}$ concentration from 11 to 19 December 2016 averaged over Beijing (blue square) and BTH (black square) region.'.

Meanwhile, we have added the definition of Beijing domain in the manuscript when we mentioned the model results for Beijing for the first time in Section 3.1 that 'The model results for Beijing in this paper are the averages over the region of blue square shown in Fig. 1a unless stated otherwise.’.

16. Fig. 3, “Beijing”. Again, Beijing point or Beijing area? Both for simulation and observation.

Response:

We have added the following sentence in the figure caption (Fig. 3): ‘The observations and simulations in Beijing were averaged over 12 observational sites and corresponding grid points, respectively.’.

17. Fig. 3: Even though the model did not consider SOA, the simulated $PM_{2.5}$ was in perfect agreement with what was observed. Is the SOA negligibly small compared to the POA during the observation period, or do the OM/OC ratio(s) assumed for OC emission in the simulation well represent those of SOA and POA in the BTH region? Specify the number of OM/OC ratio(s) used in the simulation and how the author determined the value(s).

Response:

Although the model did not consider SOA, the surface-layer $PM_{2.5}$ was overestimated by the model. The model performance in simulating $PM_{2.5}$ was described in the second paragraph of Section 3.1 that ‘The model can reasonably reproduce the temporal variations of $PM_{2.5}$ and the correlation coefficients between simulated and observed hourly concentrations are 0.77. For hourly $PM_{2.5}$, for example, the MBs (NMBs) are 29.1 μg m^{-3} (82.5%) on clean days and 6.3 μg m^{-3} (3.5%) during the two haze events. The possible reasons for the overall overestimation of $PM_{2.5}$ are as follows: (1) the model biases in underestimating WS10 and daytime PBLH; (2) the uncertainties in anthropogenic emission data (e.g. the overestimation in the BC emissions) (Qiu et al., 2017; Chen et al., 2021).’.

Gao et al. (2016) used the WRF-Chem model and showed that although the total $PM_{2.5}$ was overestimated by 43.3 μg m^{-3} (36.5%) during haze event, OC was underestimated by 44.5% due to the large uncertainty of OC emission inventory and missing secondary organic aerosol formation in the selected CBMZ-MOSAIC coupled mechanism. We also summarized the measured OC concentrations during haze days in Table R1. In this study, the mean simulated surface-layer OC concentrations was 33.0 μg m^{-3} in Beijing averaged over two haze events (mean $PM_{2.5}$ was 186.1 μg m^{-3}) shown in Fig. R1. Compared to measured OC during haze days from previous studies, the simulated OC concentrations were lower in our study.

Table R1. A summary of the measured OC concentration (μg m^{-3}) from previous studies.

Location	$PM_{2.5}$ (μg m^{-3})	OC (μg m^{-3})	Reference
Beijing, China	215.3-372.4	66.0-129.8	Gao et al., 2016
Beijing, China	150.0-250.0	22.5-37.5	Ji et al., 2017
Beijing, China	209.6	54.1	Qiu et al., 2017

Beijing, China	110.0	33.2	Chen et al., 2019
Shijiazhuang, China	216.1	79.2	Chen et al., 2019

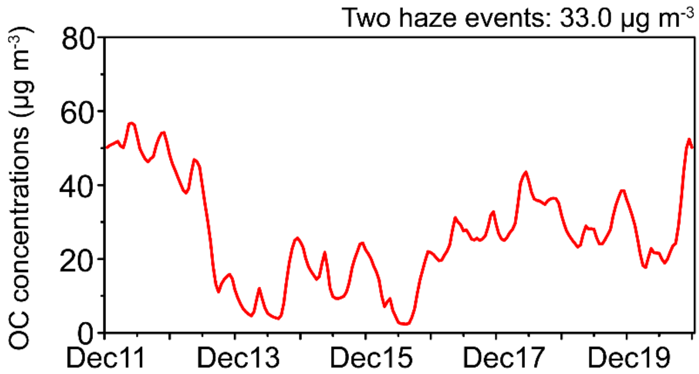

Figure R1. Time series of simulated hourly OC concentrations from 11 to 19 December 2016 averaged over Beijing. The mean value of OC averaged over the two haze events was indicated above the panel.

18. Caption of Fig. 4: What time? Both for observation and simulation.

Response:

The specific observation time was summarized in Table S1. We have also added the time of observation in Fig. 4 (on top of each panel, see below). The model results are the two-hour averages around the observation time.

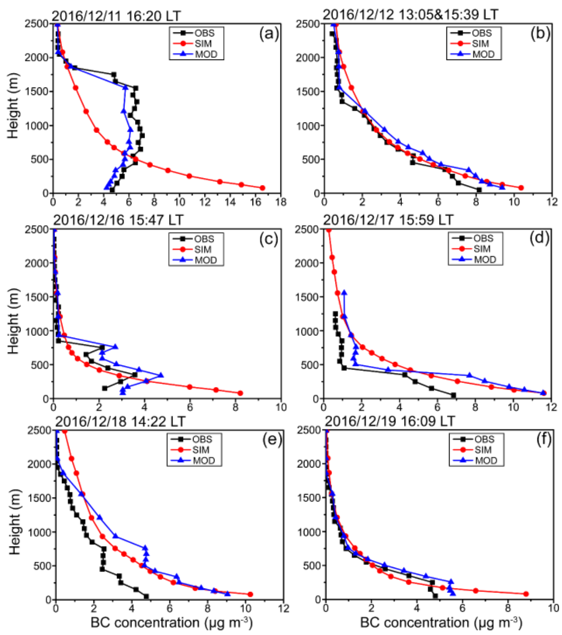

Figure 4. Observed (black line), simulated (red line) and modified (blue line) BC vertical profiles in Beijing on 11-12 and 16-19 December 2016. The time of observation is indicated on top of each panel. The model results are two-hour averages around the observation time.

19. Fig. 5: Can you compare downward solar radiation at ground surface here? It could also effectively evaluate the model performance of aerosols, and even clouds.

Response:

The comparisons between simulated and observed 3-hourly total cloud cover (%) and 3-hourly shortwave downward radiation flux (SWDOWN, W m^{-2}) in Beijing are added in Figure 5f and 5h. The corresponding statistical metrics are added in Table 3. See also our responses to your Comment #1 (Major concerns/questions).

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